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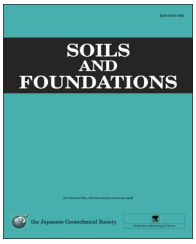

Modeling water–NAPL–air three-phase capillary behavior in soils

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Abstract

Existing models relating the capillary pressures and the degrees of saturation of NAPL, water, air three-phase are briefly reviewed, and their limitations are discussed. Of particular interest is their inability to describe the intrusion and emission behavior of NAPL precisely, especially when the degree of saturation of NAPL is low. A new variable μ representing the relative magnitude of NAPL pressure to water pressure and air pressure is then defined as a state parameter, and a novel water–NAPL–air three-phase model is proposed, which considers the transition phenomena between water–NAPL–air three-phase and arbitrary two-phase system through the parameter μ. The validity of the proposed model is checked by comparing the calculation results with those obtained by an ordinary model and existing experimental results. The proposed model is shown capable of properly considering the interactions among the water–NAPL–air three-phase, and predicting the actual retention behavior of NAPL in unsaturated ground.
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Keywords: NAPL; Unsaturated soil; Porous media; Degree of saturation; Void pressure

1. Introduction

Soil contamination occurs due to the leakage of waste materials from waste storage facilities or reservoirs. The difference of the transport mechanisms of the contaminant materials in the ground, which are dominated by their type and characteristics, has to be properly considered in addressing soil contamination problems. A number of contaminant materials, including heavy metals and agrochemicals, dissolve readily in the groundwater and transport by advection, diffusion or dispersion. There are some numerical methods for determining the seepage behavior of groundwater which are usually used to predict the movement of such water-soluble contaminants. Meanwhile, several contaminant fluids, including chlorinated solvents and petroleum products, which do not mix readily with water and which are known as non-aqueous phase liquids (NAPLs), permeate into the ground in the form of oily liquids and NAPLs, and flow differently from ground water or air. NAPLs leaked around the ground surface penetrate into the subsurface, into unsaturated ground, as schematically shown in Fig. 1. Light NAPLs (LNAPLs), such as gasoline, are less dense than water and finally float on the groundwater table. On the other hand, dense NAPLs (DNAPLs), such as the trichloroethylene solvent, are denser than water and tend to sink into the groundwater. That is, NAPLs form a complex two-phase or three-phase flow with water and/or air in the ground. Therefore, a method which can simulate water–NAPL–air three-phase seepage behavior is strongly required to cope with the problems of contamination and purification of the subsurface region by NAPLs.

In order to simulate the transport phenomena of NAPLs in unsaturated ground, a three-phase characteristic curve model

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Nomenclature

a	fitting parameter for three-phase characteristic curve
m	fitting parameters for characteristic curve proposed by van Genuchten
n	fitting parameter for characteristic curve proposed by van Genuchten
p_a	pore air pressure
p_o	pore NAPL pressure
p_w	pore water pressure
S	degree of saturation
$S_{\max}$	maximum degree of saturation of the wetting fluid
$S_{\min}$	minimum degree of saturation of the wetting fluid
S^a_{aw}	degree of saturation of air in water–air two-phase
S^w_{aw}	degree of saturation of water in water–air two-phase
S^o_{ow}	degree of saturation of NAPL in water–NAPL two-phase
S^w_{ow}	degree of saturation of water in water–NAPL two-phase
S^a_{ao}	degree of saturation of air in NAPL–air two-phase
S^o_{ao}	degree of saturation of NAPL in NAPL–air two-phase
S^a_{aow}	degree of saturation of air in water–NAPL–air three-phase
S^o_{aow}	degree of saturation of NAPL in water–NAPL–air three-phase
S^w_{aow}	degree of saturation of water in water–NAPL–air three-phase
S^l_{aow}	degree of saturation of liquid in water–NAPL–air three-phase ($=S^w_{aow}+S^o_{aow}$)
s	suction defined as the pressure difference between two void fluids
s_{aw}	suction between air and water ($=p_a-p_w$)
s_{ow}	suction between NAPL and water ($=p_o-p_w$)
s_{ao}	suction between air and NAPL ($=p_a-p_o$)
α	material parameter for characteristic curve proposed by van Genuchten
β_{aw}	scaling parameter ($=\sigma_{aw}/\sigma_{aw}=1$)
β_{ow}	scaling parameter ($=\sigma_{aw}/\sigma_{ow}$)
β_{ao}	scaling parameter ($=\sigma_{aw}/\sigma_{ao}$)
β^l	scaling parameter for three-phase characteristic curve given as a function of β_{ow} and μ
β^w	scaling parameter for three-phase characteristic curve given as a function of β_{ao} and μ
μ	relative magnitude of NAPL pressure to water pressure and air pressure ($=(p_o-p_w)/(p_a-p_w)$)
σ_{aw}	interfacial tension between air and water
σ_{ow}	interfacial tension between NAPL and water
σ_{ao}	interfacial tension between air and NAPL

giving the capillary pressure–saturation relationship of water, NAPL and air phase is crucial. In past research, soil water characteristic curve models for water–air capillary relationship of unsaturated soils have been proposed by Brooks and Corey (1966), van Genuchten (1980) and some other researchers, and these models are commonly used in practical applications due to their simplicity. Parker et al. (1987) investigated a two-phase retention behavior in water–NAPL and NAPL–air, and claimed that arbitrary two-phase capillary phenomena can be described by scaling up or down the water–air two-phase characteristic curve such as one proposed by van Genuchten along the suction axis. The applicability of this method was checked through the comparison with the experimental results by Liu et al. (1998). Meanwhile, Parker and Lenhard (1990) investigated the pressure–saturation relationship of the water–NAPL–air three-phase system and proposed a model for the three-phase capillary relationship. In this model, the water and liquid saturations are given by a two-phase characteristic curve for the water–NAPL system and that for the NAPL–air system separately as NAPL, which has a medium wettability, always exists between the water and air phase according to the assumption by Leverett (1941). Lenhard and Parker (1988) checked the applicability of the model by comparing with the experimental evidence. The hysteretic capillary behavior of water, NAPL and air has also been investigated. Land (1968) discussed hysteresis in two-phase system, and Parker and Lenhard (1987a, 1987b), Lenhard et al. (1989) and Lenhard (1992) investigated the hysteresis effect in the water–NAPL–air three-phase system. Moreover, several researchers have applied the characteristic curve model for the three-phase system to initial value problems of soil contamination and remediation.

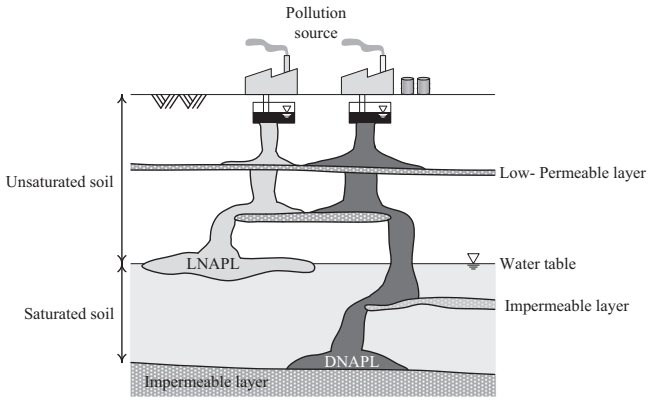

Fig. 1. Schematic figure of contamination of unsaturated shallow ground by LNAPL and DNAPL.

Although the water–NAPL–air three-phase characteristic curve model behavior proposed by Parker and Lenhard (1990) is certainly simple and tends to be applied to the simulation of multiphase flow problems in the ground, Hofstee et al. (1997) stated that, due to its comparatively low broadening coefficient, this model cannot describe the transport phenomena of NAPL. In addition, this model cannot consider the transition to the water–air two-phase relationship because it depends on a premise that NAPL exists, as was pointed out by

van Dijke and van der Zee (1997) and the penetration process of NAPL into unsaturated ground or the remediation process of NAPL from water–NAPL–air three-phase system cannot consequently be simulated properly by this model. Based on their experimental results, Tanahashi et al. (2006) pointed out that the model developed by Parker and Lenhard (1990) cannot predict the capillary behavior of the water–NAPL–air three-phase system in soils when the NAPL saturation is low. Therefore, in this paper, a rational characteristic curve model for water–NAPL–air three-phase capillary behavior in soils which suitably incorporates transition phenomena between the three-phase and arbitrary two-phase system is proposed in order to overcome the limitations of the existing models.

2. Overview of a model for two-phase characteristic curve

Several characteristic curve models have been proposed for unsaturated soils whose void consists of water and air, and one by van Genuchten (1980) is usually used due to its simplicity. In this model, degree of saturation S is given as a function of suction s, pressure difference of two fluids, in the following form.

$$S = S(s) = S_{\min} + (S_{\max} - S_{\min})\{1 + (\alpha s)^n\}^{-m} \quad (1)$$

The material parameter α has a dimension of reciprocal pressure. van Genuchten (1980) proposed an empirical relationship between non-dimensional fitting parameters m and n that $m = 1 - 1/n$. The relationship between the degree of water saturation and suction in unsaturated soil is given by Eq. (1) as

$$S^w_{aw} = S(s_{aw}) = S_{\min} + (S_{\max} - S_{\min})\{1 + (\alpha s_{aw})^n\}^{-m} \quad (2)$$

In this paper, superscripts represent the void fluid indicated by the symbol and subscripts represent the combination of void fluids and w, a and o represents water, air and NAPL (oil) phase, respectively. Eq. (1) obviously gives a unique relationship between suction and degree of saturation. In this regard, Gardner (1958), Brooks and Corey (1966), Fredlund and Xing (1994) and others proposed similar models. In water–air two-phase system, Eq. (2) always holds in an air phase.

$$S^a_{aw} = 1 - S^w_{aw} \quad (3)$$

Parker et al. (1987) applied this model to describe the retention behavior of the water–NAPL or NAPL–air two-phase in soils. Among two void fluids, the degree of saturation of void fluid with a higher wettability is given by Eq. (1). Wettability is defined as the inverse of the contact angle, which is the angle at which the two-phase interface meets on the solid surface as indicated by Fig. 2. Void fluids with an acute angle at the two-phase fluid interface have a higher wettability. Although wettability depends on the kind of NAPL, water usually has the highest wettability, and is followed by NAPL and the air in this order. Thus, in the water–NAPL two-phase system and NAPL–air two-phase system, the degree of water saturation and that of NAPL saturation are given in a similar way as Eq. (1), respectively.

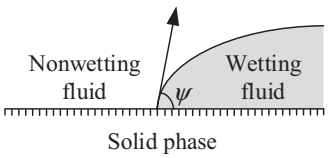

Fig. 2. Wettability and contact angle ψ.

$$S^w_{ow} = S(\beta_{ow} s_{ow}) = S_{\min} + (S_{\max} - S_{\min})\{1 + (\alpha\beta_{ow} s_{ow})^n\}^{-m} \quad (4)$$

$$S^o_{ao} = S(\beta_{ao} s_{ao}) = S_{\min} + (S_{\max} - S_{\min})\{1 + (\alpha\beta_{ao} s_{ao})^n\}^{-m} \quad (5)$$

The scaling parameters β_{ow} and β_{ao} are defined as the ratio of interfacial tension σ_{aw} between air and water to that between the focused two-phase fluids as follows.

$$\beta_{ow} = \sigma_{aw}/\sigma_{ow} \quad (6)$$

$$\beta_{ao} = \sigma_{aw}/\sigma_{ao} \quad (7)$$

For water–air two-phase system, scaling parameter β_{aw} is obviously equal to 1 and is already considered in Eq. (2). The validity of the scaling parameter β is confirmed through experiments by Parker et al. (1987) and Liu et al. (1998). For the above equations, degrees of saturation of void fluid having lower wettability are given by Eqs. (8) and (9), respectively.

$$S^o_{ow} = 1 - S^w_{ow} \quad (8)$$

$$S^a_{ao} = 1 - S^o_{ao} \quad (9)$$

It is indicated from Eqs. (2), (4) and (5) that capillary behavior of an arbitrary two-phase fluid system in soils can be described in a consistent manner by scaling down the water–air characteristic curve by β along the suction axis.

3. Overview of a model for three-phase water–NAPL–air characteristic curve

In the water–NAPL–air three-phase system in soils, arbitrary three relationships of water–air, water–NAPL and NAPL–air system may possibly exist and would be given in a similar form as Eqs. (2), (4) and (5). In addition, Eq. (10) has to always be satisfied.

$$S^w_{aow} + S^o_{aow} + S^a_{aow} = 1 \quad (10)$$

The subscript aow represents the water–NAPL(oil)–air three-phase system. As three values – the water, NAPL and air saturations – all four Eqs. (2), (4), (5) and (10) cannot be satisfied due to the mathematical inconsistency as the number of equations exceeds the number of unknowns by one.

Parker and Lenhard (1990) proposed a model for the three-phase saturation–suction relationship in which the number of conditional equations is reduced by one. This model is formulated based on the assumption which is originally proposed by Leverett (1941): the void fluid with the higher wettability tends to reside in the proximity of the contact point of soil particles as shown in Fig. 3. In this scenario, NAPL always exists in between the water phase and air phase as it has intermediate wettability. This assumption means that the water saturation is given by a unique function of the capillary pressure across the water–NAPL

interface only, and the total liquid saturation is defined by a unique function of the capillary pressure across the NAPL–air interface. Consequently, Eq. (2) is not considered as water and air phases, but are assumed to have no contact with each other in the three-phase system. Based on this assumption, Eqs. (11) and (12) are applied in addition to Eq. (10).

$$S^w_{aow} = S^w_{ow} = S(\beta_{ow}s_{ow}) = S_{\min} + (S_{\max} - S_{\min})\{1 + (\alpha\beta_{ow}s_{ow})^n\}^{-m} \tag{11}$$

$$S^l_{aow} = S^o_{ao} = S(\beta_{ao}s_{ao}) = S_{\min} + (S_{\max} - S_{\min})\{1 + (\alpha\beta_{ao}s_{ao})^n\}^{-m} \tag{12}$$

Here, superscript l represents the liquid phase consisting of water and NAPL. In this model, the water and liquid saturations are given separately by van Genuchten's equation, and the interaction between the water and air phases is not considered as water–air suction s_{aw} is not included in Eqs. (11) and (12). The three-phase characteristic curve model proposed by Parker and Lenhard (1990) is called the PL model for short hereafter.

The degree of liquid saturation S^l_{aow} representing the summation of the degrees of water and NAPL saturation must be larger than the degree of water saturation S^w_{aow}.

$$S^l_{aow} - S^w_{aow} = S^o_{aow} \geq 0 \tag{13}$$

However, Eq. (13) is not necessarily satisfied in the PL model as the degree of water saturation S^w_{aow} is given by Eq. (11) irrespective of Eq. (12) giving degree of liquid saturation S^l_{aow}. Therefore, in the PL model, S^w_{aow} and S^l_{aow} are both assumed to be equal to the water saturation S^w_{ao} given by Eq. (2) as a temporal expedient when S^w_{aow} exceeds S^l_{aow}. The capillary relationship of the PL model is summarized as follows.

$$\begin{cases} S^l_{aow} = S(\beta_{ao}s_{ao}),\ S^w_{aow} = S(\beta_{ow}s_{ow}) & \text{when } S(\beta_{ao}s_{ao}) \geq S(\beta_{ow}s_{ow}) \\ S^l_{aow} = S^w_{aow} = S(s_{aw}) & \text{when } S(\beta_{ao}s_{ao}) < S(\beta_{ow}s_{ow}) \end{cases} \tag{14}$$

Fig. 4 shows examples of the pressure–saturation relationships of water, NAPL and air phases calculated by the PL model. The material parameter used in the calculation is summarized in Table 1. The dotted lines in this figure show the two-phase capillary relationships given by Eqs. (2), (11) and (12). In figure (a), only the NAPL pressure p_o is varied under constant water pressure p_w of 20 kPa and a constant air pressure p_a of 50 kPa. As shown in this figure, the capillary relationship becomes awkwardly discontinuous due to the limitation of the LP model when the NAPL saturation becomes zero and the capillary relationship shifts to that of the water–air two-phase system. It is also known from figure (a) that the

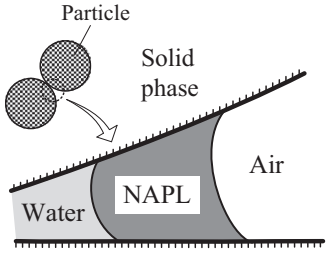

Fig. 3. Assumption by Leverett on the void fluid around the contact point of soil particle.

Table 1
Parameters for the characteristic curve model.

$S_{\max}$	1.0
$S_{\min}$	0.0
α	0.038
β_{aw}	1.0
β_{ow}	1.77
β_{ao}	2.76
n	2.91
m	0.656

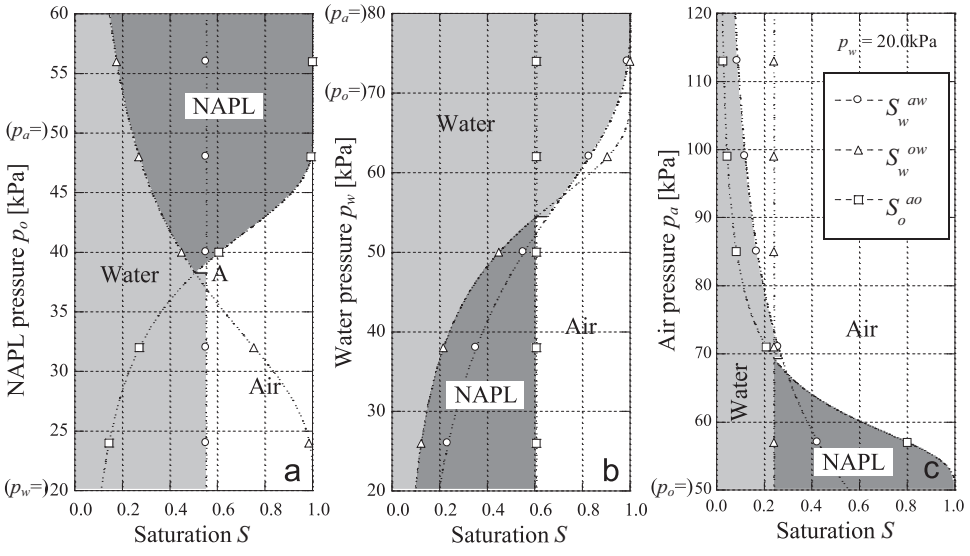

Fig. 4. Variations in degrees of saturation of water, NAPL and air calculated by an existing model proposed by Parker and Lenhard (the PL model). (a) NAPL pressure variation under constant water pressure p_w (=20 kPa) and constant air pressure p_a (=50 kPa), (b) water pressure variation under constant NAPL pressure p_o (=70 kPa) and constant air pressure p_a (=80 kPa) and (c) air pressure variation under constant water pressure p_w (=20 kPa) and NAPL pressure p_o (=50 kPa).

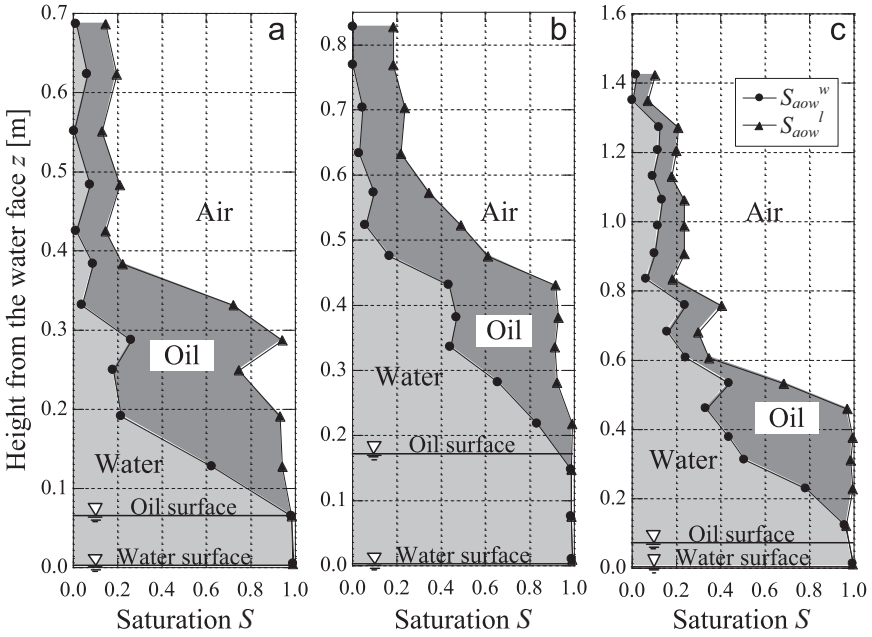

Fig. 5. Vertical distributions of degree of saturation of water, NAPL and air observed in gravity drainage column experiments using (a) aggregated glass beads having diameter of 0.4 mm, (b) aggregated glass beads having diameter of 0.2 mm and (c) Toyoura sand (replotted from Tanahashi et al. (2006)).

degree of saturation of each phase becomes constant even though the NAPL pressure is varied under the threshold NAPL pressure. In figure (b), the water pressure p_w varies under constant NAPL pressure p_o of 70 kPa and constant air pressure p_a of 80 kPa, while the responses of the saturations of each phase due to the variation in air pressure p_a is checked under constant p_w of 20 kPa and constant p_o of 50 kPa in figure (c). In figure (b) it can be seen that increases in water pressure only decrease NAPL saturation and have no influence on air saturation in the presence of NAPL. It can also seen in figure (c) that variations in air pressure only affect the degree of NAPL saturation and do not change the water saturation in the three-phase system. These results are obviously not consistent with actual phenomena and can be explained by the premise that the NAPL phase is persistent in the PL model and does not consider interaction between the water and air phases.

In Fig. 4 it can be seen that the degrees of water and air saturation become discontinuous at the boundary between the three-phase and water–air two-phase where the NAPL saturation becomes zero. This is because Eqs. (11) and (12) cannot satisfy Eq. (13) and the degree of water saturation calculated by these equations differs from that given by Eq. (2). In order to avoid such discontinuity, capillary curves by Eqs. (2), (11) and (12) must intersect at one point. At the intersection point, Eq. (15) holds.

$$\beta_{aw}s_{aw} = \beta_{ow}s_{ow} = \beta_{ao}s_{ao} \tag{15}$$

Now, suctions s, pressure differences of two fluids among water, NAPL and air phase satisfy the following identity.

$$s_{aw} = p_a - p_w = (p_a - p_o) + (p_o - p_w) = s_{ao} + s_{ow} \tag{16}$$

By substituting Eq. (15) for Eq. (16), relationship between scaling coefficients β for the combinations of two fluids is obtained.

$$\frac{1}{\beta_{aw}} = \frac{1}{\beta_{ow}} + \frac{1}{\beta_{ao}} \tag{17}$$

By substituting the surface tensions of each phase, Eq. (18) is obtained.

$$\sigma_{aw} = \sigma_{ow} + \sigma_{ao} \tag{18}$$

This equation suggests that the discontinuity of water and air saturations can be avoided only when the surface tension of NAPL satisfies Eq. (18).

Tanahashi et al. (2006) discussed the applicability of the PL model through laboratory tests and theoretical investigation. They performed vertical gravity drainage column experiments, in which surfaces of water and kerosene (NAPL) were controlled at the lower part of the soil column and distributions of the degrees of saturation of water and NAPL above the water surface were carefully measured. Three kinds of materials were used in the model ground: aggregates of glass beads with a diameter of 0.2 mm, aggregates of glass beads with a diameter of 0.4 mm and Toyoura sand. Fig. 5 shows the vertical distributions of the degrees of saturation of water, NAPL observed in their experiments. They indicated that degree of water saturation gradually decreases in the region above the water surface and NAPL thinly cumulates on the water layer. Meanwhile, Fig. 6 shows our corresponding calculation results using the LP model, in which black plots indicates the corresponding experimental values by Tanahashi et al. (2006). The vertical distributions of capillary pressures of water, NAPL and air assumed in the calculation are schematically shown in Fig. 7. The capillary pressures of water and NAPL were assumed to be zero at their respective surfaces, and their distributions were assumed to be linear with an

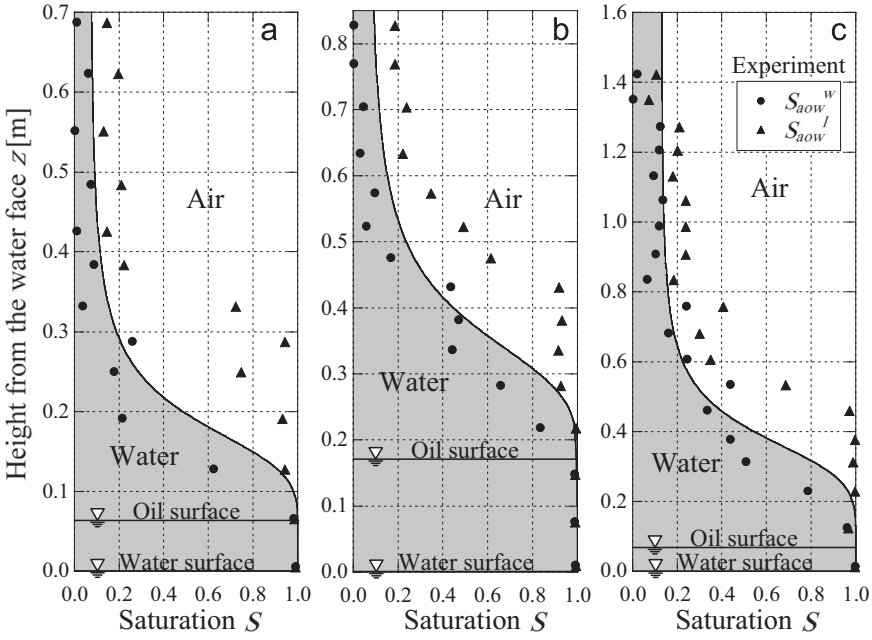

Fig. 6. Simulations of gravity drainage column experiments by an existing model proposed by Parker and Lenhard (the PL model). (a) Aggregates of glass beads having diameter of 0.4 mm, (b) aggregates of glass beads having diameter of 0.2 mm and (c) Toyoura sand.

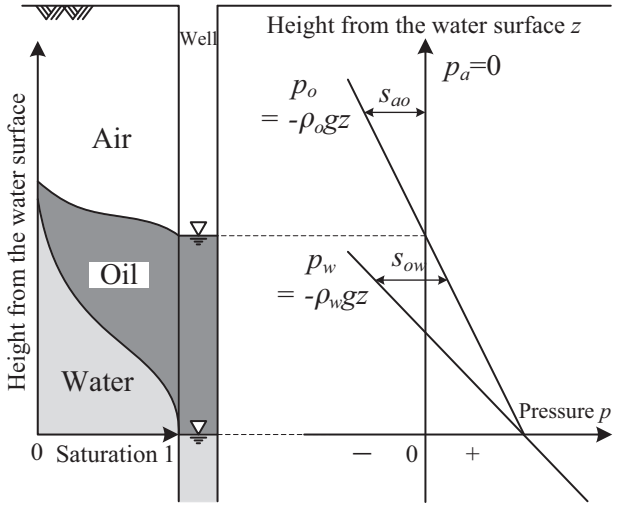

Fig. 7. Outline of the gravity drainage column experiments (Tanahashi et al., 2006) and assumed pressure distributions.

Table 2
Physical properties of water–NAPL–air three-phase fluids used in the experiments.

σ_{aw} [10^{-3} N/m]	72.75
σ_{ow} [10^{-3} N/m]	41.0
σ_{ao} [10^{-3} N/m]	26.4
ρ_0 [g/cm^3]	0.8

inclination of their densities. The physical properties of kerosene are listed in Table 2. Parameters for the PL model, which are determined by fitting calculation results with the observed NAPL–air characteristic curves shown in Fig. 8, are listed in Table 3. Both the gravity drainage column experiments and their corresponding simulations using the PL model show that water thickly cumulates in the region above the water surface and degree of water saturation gradually decreases to zero. It is clearly seen that the PL model cannot predict the formation of a thin layer of NAPL in region above the NAPL surface observed in the experiments. As was already discussed with regard to Fig. 4, the disadvantages of the PL model are that the variations of degrees of saturation of water, NAPL and air predicted by this model have unnatural discontinuities and that the NAPL saturation becomes zero when the NAPL pressure becomes relatively small. This explains why the LP model cannot predict the existence of NAPL when the level of the NAPL surface is rather close to the water level.

Although an existing three-phase characteristic curve model developed by Parker and Lenhard (1990) is certainly simple and widely used in simulating multiphase seepage behavior in the ground, the prediction accuracy of the model is rather poor when the degree of NAPL saturation is relatively low. In the field of geoenvironmental engineering or contaminant hydrology, the penetration of NAPLs into the water–air two-phase system and purification of NAPLs from water–NAPL–air three-phase system are quite important issues and a three-phase capillary model incorporating the transition between the water–air two-phase system and the water–NAPL–air three-phase system is strongly expected. Therefore, a model which overcomes these problems with existing models and which improves its accuracy is proposed in this paper.

4. A state variable for water–NAPL–air three-phase capillary relationship and interpretation of an existing model for three-phase

Our aim is to develop a model which suitably incorporates the intrusion and discharge behavior of NAPL by taking

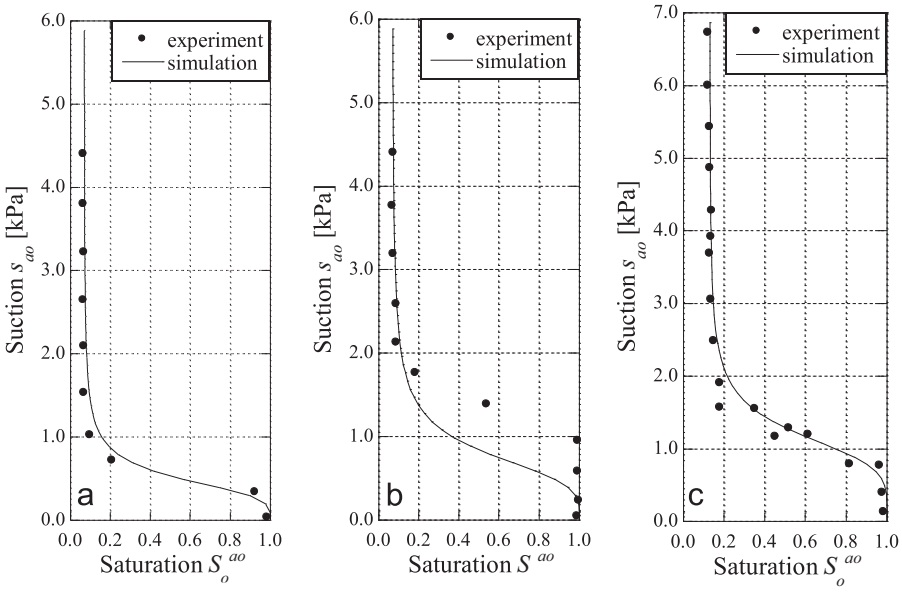

Fig. 8. Relationships between capillary pressure and saturation of NAPL and air in two-phase system (Tanahashi et al., 2006) and its calculation by two-phase relationship. (a) Glass beads 0.4 mm, (b) glass beads 0.2 mm and (c) Toyoura sand.

Table 3
Parameters for an existing model proposed by Parker and Lenhard (the PL model) for three kinds of materials.

	Glass beads 0.4 mm	Glass beads 0.2 mm	Toyoura sand
S_{max}	1.0	1.0	1.0
S_{min}	0.07	0.07	0.13
α	0.79	0.50	0.32
β_{aw}	1.0	1.0	1.0
β_{ow}	1.77	1.77	1.77
β_{ao}	2.76	2.76	2.76
n	4.0	4.0	5.0
m	0.75	0.75	0.80

account of the interaction between the water and air phases. It is reasonable to assume the following conditions in the proposed model: the NAPL phase is swept by the water phase and its saturation S^o_{aow} becomes zero when NAPL pressure p_o is equal to the water pressure p_w and the suction s_{aw} between air and water becomes zero; the water–NAPL–air three phases exist when p_o is larger than p_w and is less than the air pressure p_a; the air phase is swept by the NAPL phase and the air saturation S^a_{aow} becomes zero when p_o is equal to p_a and suction s_{ao} between air and NAPL becomes zero. In order to grasp the above-mentioned transitions, a new variable μ representing a relative magnitude of NAPL pressure p_o to water pressure p_w and air pressure p_a is employed here as a state variable.

$$\mu = \frac{p_o - p_w}{p_a - p_w}\left(= \frac{s_{ow}}{s_{aw}}\right) \tag{19}$$

μ becomes 0 when the NAPL pressure coincides with the air pressure and μ becomes 1 when the NAPL pressure coincides with the water pressure. Transition of the void fluids is summarized simply through the state variable μ as follows.

$$\mu = 0\ (p_w = p_o < p_a) : \text{water} - \text{air two} - \text{phase state}$$
$$0 < \mu < 1\ (p_w < p_o < p_a) : \text{water} - \text{NAPL} - \text{air three} - \text{phase state}$$
$$\mu = 1\ (p_w < p_o = p_a) : \text{water} - \text{NAPL two} - \text{phase state} \tag{20}$$

Suctions s_{ow} and s_{ao} can be related with s_{aw} by μ as

$$s_{ow} = \mu s_{aw} \tag{21}$$

$$s_{ao} = (1-\mu)s_{aw} \tag{22}$$

Characteristic of the PL model is investigated here by substituting Eqs. (21) and (22) into Eq. (14).

$$\begin{cases} S^l_{aow} = S(\beta_{ao}(1-\mu)s_{aw}) \text{ and } S^w_{aow} = S(\beta_{ow}\mu s_{aw}) & \text{when } S(\beta_{ao}(1-\mu)s_{aw}) \geq S(\beta_{ow}\mu s_{aw}) \\ S^l_{aow} = S^w_{aow} = S(s_{aw}) & \text{when } S(\beta_{ao}(1-\mu)s_{aw}) < S(\beta_{ow}\mu s_{aw}) \end{cases} \tag{23}$$

Note that three-phase capillary pressure–saturation relationship is described uniquely by a single suction s_{aw} in Eq. (23). This equation can be rewritten quite concisely as follows.

$$S^l_{aow} = S(\beta^l s_{aw}),\quad S^w_{aow} = S(\beta^w s_{aw})$$

$$\text{where } \begin{cases} \beta^l = \beta_{ao}(1-\mu) \text{ and } \beta^w = \beta_{ow}\mu & \text{when } \beta^l \leq \beta^w \\ \beta^l = \beta^w = 1\ (=\beta_{aw}) & \text{when } \beta^l > \beta^w \end{cases} \tag{24}$$

Here, β^l and β^w are given as functions of β_{ao}, β_{ow} and the state variable μ and they control the three-phase characteristic curves of liquid and water. Variations of β^l and β^w in the PL model are shown in Fig. 9, where the values of β_{ao} and β_{ow} listed in Table 1 are applied. It is known from this figure that both β^l and β^w are equal to 1 ($=\beta_{aw}$) when μ is equal to 0 and that β^w increases to β_{ow} when μ increases to 1. Such relationships are essential in describing the transition of the three-phase system to water–air or water–NAPL two-phase system.

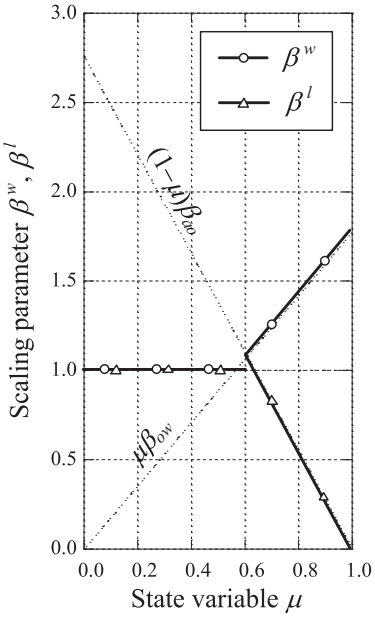

Fig. 9. Variations of scaling parameters β^l and β^w in an existing model proposed by Parker and Lenhard (the PL model).

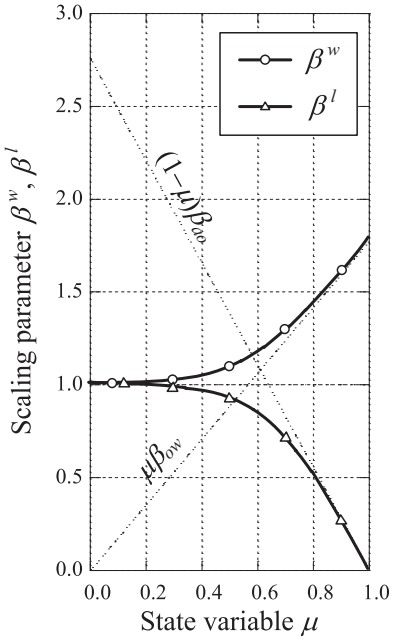

Fig. 10. Variations of scaling parameters β^l and β^w in the proposed model.

However, as could be seen in Fig. 6, both β^l and β^w are discontinuous bilinear functions of μ and their discontinuities causes the discontinuities of liquid and water saturations. In addition, the NAPL phase cannot exist in the region where μ is less than $0.61\left(=\beta_{ao}/(\beta_{ao}+\beta_{ow})\right)$, which would be inconsistent with actual phenomena.

5. A model for three-phase water–NAPL–air characteristic curve considering intrusion and discharge process of NAPL phase

An outline of the proposed model for three-phase characteristic curve, which incorporates smooth transition between three-phase and arbitrary two-phase system, is explained here. In the proposed model, liquid and water saturations are given in the similar way as the PL models as follows:

$$S^l_{aow}=S\left(\beta^l(\mu)s_{aw}\right),\quad S^w_{aow}=S(\beta^w(\mu)s_{aw}) \tag{25}$$

The state variable μ, the relative magnitude of NAPL pressure given by Eq. (19), works as an intermediary in the proposed model. Based on the discussions in the previous section, β^l and β^w have to be functions of μ satisfying the following requirements.

$$\begin{cases}\beta^l(0)=\beta^w(0)=1\left(=\beta_{aw}\right)\\ \beta^l(1)=0,\ \ \beta^w(1)=\beta_{ow}\\ \beta^l(\mu)\le\beta^w(\mu)\end{cases} \tag{26}$$

Although any function satisfying the above-mentioned requirements can be applied for β^l and β^w, a quadratic Bezier curve (see Appendix A) was applied to describe β^l and β^w in the present study. Variations of β^l and β^w with μ are shown in Fig. 10, where values of β_{ao} and β_{ao} used are listed in Table 1 and fitting parameter a for Bezier curve applied here is 2.67. β^l is a monotonously decreasing continuous function which smoothly connects two linear lines given by $\beta^l=\beta_{ao}(1-\mu)$ and $\beta^l=1\ \left(=\beta_{aw}\right)$, while β^w is a monotonically increasing, continuous function connecting $\beta^w=\beta_{ow}\mu$ and $\beta^w=1\ \left(=\beta_{aw}\right)$.

Calculations of capillary pressures–saturations relationships are rather simple. Incremental relationships between degrees of saturation and pressures are given as follows.

$$dS^l_{aow}=\frac{\partial S}{\partial\beta^l}\,d\beta^l+\frac{\partial S}{\partial s_{aw}}\,ds_{aw},\quad dS^w_{aow}=\frac{\partial S}{\partial\beta^w}\,d\beta^w+\frac{\partial S}{\partial s_{aw}}\,ds_{aw} \tag{27}$$

β^l and β^w are functions of μ.

$$d\beta^l=\frac{\partial\beta^l}{\partial\mu}\,d\mu,\quad d\beta^w=\frac{\partial\beta^w}{\partial\mu}\,d\mu \tag{28}$$

μ and s_{aw} are functions of fluid pressures.

$$d\mu=\frac{\partial\mu}{\partial p_w}\,dp_w+\frac{\partial\mu}{\partial p_o}\,dp_o+\frac{\partial\mu}{\partial p_a}\,dp_a,\quad ds_{aw}=dp_a-dp_w \tag{29}$$

The applicability of the proposed model is firstly investigated through comparisons with the results calculated by the PL model. Fig. 11 shows the results of the calculations by the proposed model, where calculations corresponding to Fig. 4 were performed with the value of the fitting parameter a of 2.67. In figures (a), (b) and (c), NAPL, and the water and air pressures are varied respectively while other fluid pressures are kept constant. It can be seen in Fig. 11 that the proposed model properly describes the gradual decrease of the degree of NAPL saturation when the water or air pressure increases or the NAPL pressure decreases, while the discontinuity in the degree of saturation is seen in case of the simulation by the PL model.

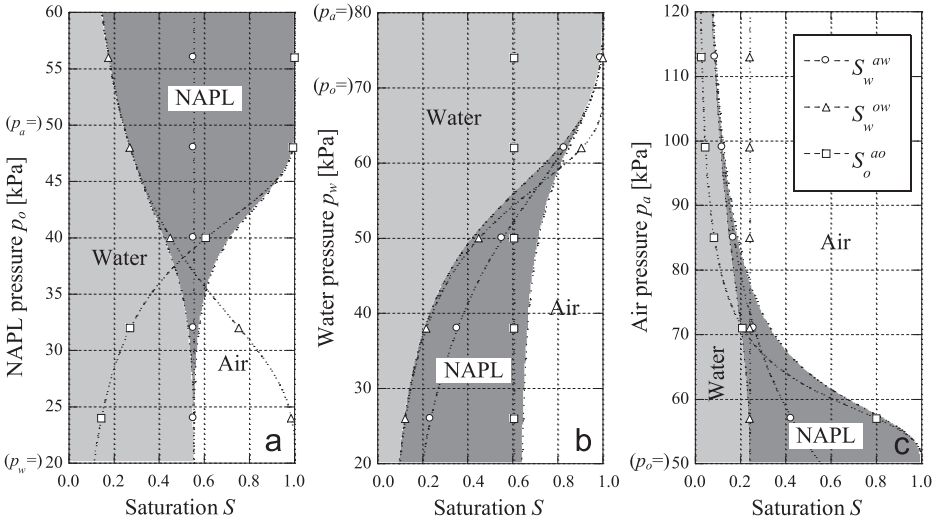

Fig. 11. Variations in degrees of saturation of water, NAPL and air calculated by the proposed model. (a) NAPL pressure variation under constant water pressure p_w (=20 kPa) and constant air pressure p_a (=50 kPa), (b) water pressure variation under constant NAPL pressure p_o (=70 kPa) and constant air pressure p_a (=80 kPa) and (c) air pressure variation under constant water pressure p_w (=20 kPa) and NAPL pressure p_o (=50 kPa).

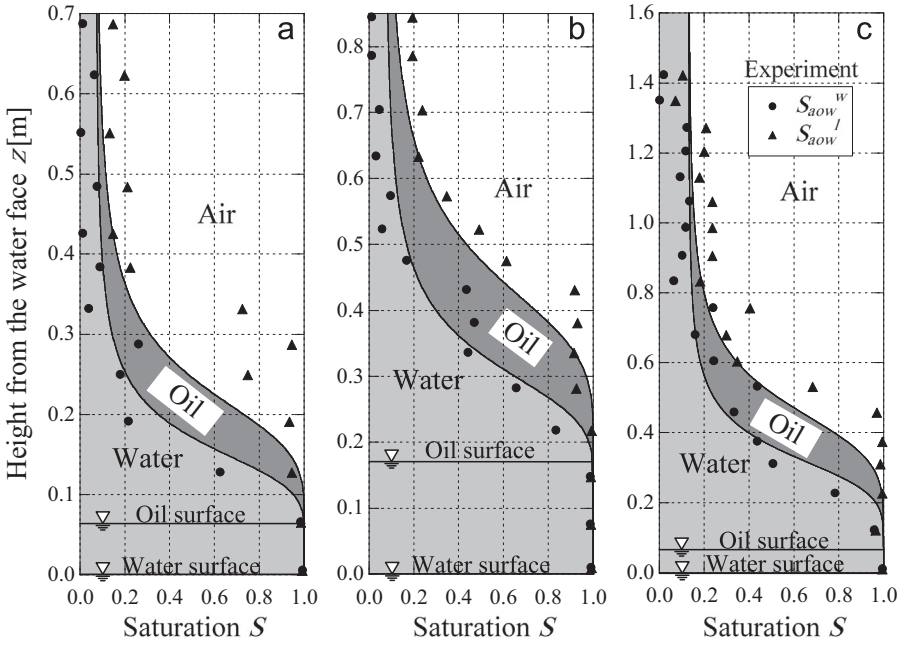

Fig. 12. Simulations of gravity drainage column experiments by the proposed model. (a) Aggregates of glass beads having diameter of 0.4 mm, (b) aggregates of glass beads having diameter of 0.2 mm and (c) Toyoura sand.

Table 4
Parameters for the proposed characteristic model for three kinds of granular materials.

	Glass beads 0.4 mm	Glass beads 0.2 mm	Toyoura sand
S_{max}	1.0	1.0	1.0
S_{min}	0.07	0.07	0.13
α	0.79	0.50	0.32
β_{aw}	1.0	1.0	1.0
β_{ow}	1.77	1.77	1.77
β_{ao}	2.76	2.76	2.76
n	4.0	4.0	5.0
m	0.75	0.75	0.80
α	−1.96	−1.96	−1.96

The validity of the proposed model is also discussed by comparing the results calculated by the proposed model with those of the gravity drainage column tests conducted by Tanahashi et al. (2006) and those obtained by the PL model. Experimental results obtained using three kinds of granular materials were already shown in Fig. 5, while their corresponding calculated results by the PL model were shown in Fig. 6. The vertical distributions of the degrees of saturation of water, NAPL (kerosene) and air in the ground calculated by the proposed are shown in Fig. 12. In each figure, closed plots represent the experimental values. Material parameters of the proposed model are listed in Table 4. Most of the parameters are common to the proposed model and the PL model. The only parameter added to the proposed model is a, which is a fitting parameter for a quadratic Bezier curve relating the value of β^l and β^w with μ. Figs. 5 and 6 indicate that, although NAPL is spread thinly above the NAPL surface in the experiments, the PL model cannot predict such a thin layer. In contrast to the PL model, the proposed model properly predicts the rather thin distributions of NAPL in unsaturated soils.

6. Conclusions

Existing soil characteristic models describing the variations of degrees of water–NAPL–air three-phase saturations due to their pressure variations are briefly reviewed and their limitations are summarized. The main issue in the modeling is that existing models cannot describe the intrusion and emission behavior of NAPL properly as they postulate the existence of NAPL and they do not take the transition between the water–air two-phase and water–NAPL–air three-phase into consideration. Therefore, even though existing characteristic models, such as that proposed by Lenhard and Parker, consider the water–air two-phase behavior separately by applying a well-known equation proposed by van Genuchten, the degree of saturation becomes awkwardly discontinuous. As a result, water–NAPL–air three-phase seepage behavior cannot be simulated precisely, and this is especially problematic when the degree of NAPL saturation is low.

In this paper, a new three-phase characteristic model incorporating the transition between an arbitrary two-phase and water–NAPL–air three-phase is proposed based on a state variable μ which represents the relative magnitude of NAPL pressure to the pressures of water and air. The proposed model considers the interaction between water and air, which is not considered in existing models, and was shown to be capable of predicting the retention behavior of water, NAPL and air well. It can also predict the disappearance of the NAPL phase and behavior when the degree of NAPL saturation is low.

Appendix A

Quadratic Bezier curve shown in Fig. A1 is described with three control points r_1, r_2 and r_3 and it has the following properties: (a) the curve interpolates r_1 and r_3 and its curvature is controlled by the middle control point r_2; (b) the curve

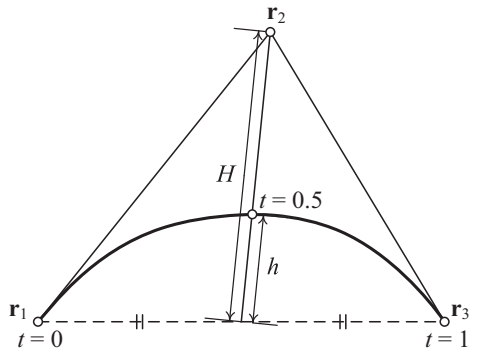

Fig. A1. Quadratic Bezier curve with three control points..

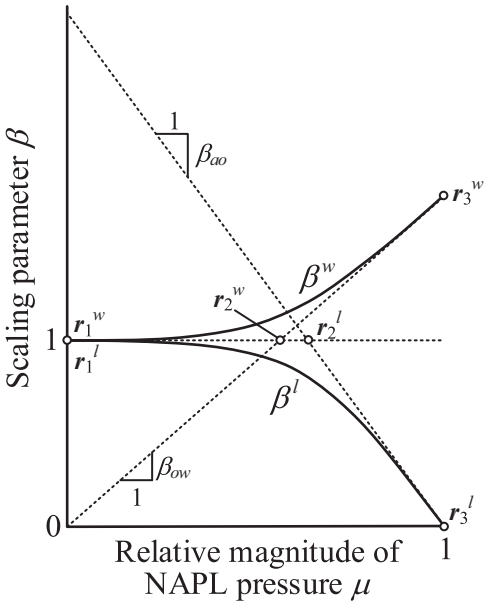

Fig. A2. Description of μ–β^l and μ–β^w relationships by quadratic Bezier curves..

connects to r_1 and r_3 smoothly, that is, tangential vector of the curve is parallel to r_1–r_2 and r_2–r_3 at r_1 and r_3, respectively. Point r on the Bezier curve can be derived via a geometric construction and can be easily written as a linear combination of the control points as

$$r = k_1 r_1 + k_2 r_2 + k_3 r_3 \tag{A1}$$

where k_1, k_2 and k_3 are functions of intermediate parameter t $(0 \le t \le 1)$ as follows.

$$\begin{cases} k_1 = (1-t)^2 \\ k_2 = 2t(1-t) \\ k_3 = t^2 \end{cases} \tag{A2}$$

In this study, quadratic Bezier curve is revised to be able to control its curvature with an additional parameter a (≥ -2) and is applied to describe β^l–μ and β^w–μ relationships. The revised representation of the curve is given as

$$r = \frac{k_1 r_1 + k_2 r_2 + k_3 r_3}{k_0} \tag{A3}$$

where

$$\begin{cases} k_1 = (1-t)^2 \\ k_2 = (2+a)t(1-t) \\ k_3 = t^2 \\ k_0 = k_1 + k_2 + k_3 \end{cases} \tag{A4}$$

Position of the passing point at $t=0.5$ is controlled by the parameter a as Eq. (A5).

$$\frac{h}{H} = \frac{2+a}{4+a} \tag{A5}$$

As shown in Fig. A2, control points r_1^w, r_2^w and r_3^w are used to describe the μ–β^w relationship and r_1^l, r_2^l and r_3^l are used for the μ–β^l relationship.

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